Ultrafast Manipulation of Spins in Antiferromagnetic Materials: New Avenue in Magnetic Memory?

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The study of ultrafast (10^{-12} s and faster) magnetization dynamics and the search for high speed magnetic switching have become topics of ultimate interest in both condensed matter physics and information storage technology.[1)-10)] So far, most studies of ultrafast spin dynamics were focused on ferromagnetic materials because of their obvious relevance for applications. However, due to the fact that in antiferromagnets no angular momentum is associated with the order parameter, spin dynamics in these materials is intrinsically much faster than in ferromagnets.[11)] Thus, in the search for the ultimate speed of magnetic dynamics and switching, one should focus attention to antiferromagnets. Here we describe our recent finding that ultrafast laser pulses can change the magnetic anisotropy of an antiferromagnet within 1 ps.[12)] This allows one to manipulate the spins of antiferromagnets on a time scale of a few picoseconds, in contrast to the hundreds of picoseconds in a ferromagnet.[6),7)] Our findings can expand the now limited set of applications of antiferromagnetic materials and revolutionize the technology of ultrafast magnetic recording.

Key words: ultrafast spin dynamics, antiferromagnets, magnetic recording

1. Introduction

All magnetically-ordered materials can be divided into two primary classes: ferromagnets and antiferromagnets. Ferromagnets are characterized by parallel alignment of their spins and posses a spontaneous magnetization. Due to this magnetization ferromagnetic materials are known from ancient time and found their application long time ago from the compass to computer storage. Antiferromagnetic materials are characterized by antiparallel alignment of their spins. Although antiferromagnets represent the overwhelming majority of magnetically ordered materials in nature, for a long time they remained an issue of academic interest only. Nevertheless, antiferromagnets exhibit much faster spin dynamics in contrast to ferromagnetic materials. For instance, to deflect the magnetization of a ferromagnet from its equilibrium, a critical field $H_{FM} \sim H_A$ of the order of the effective anisotropy field H_A is required. In contrast, the response of an antiferromagnet to an applied field remains very weak until the critical field $H_{AFM} \sim \sqrt{(H_A H_{ex})}$ is reached. In most materials the exchange field $H_{ex} \gg H_A$, and thus $H_{AFM} \gg H_{FM}$. This large rigidity also shows up in the magnetic resonance. The frequency of the homogeneous mode of spin excitations in antiferromagnets is equal to $\omega = \gamma\sqrt{(H_A H_{ex})}$. This is in contrast to $\omega = \gamma H_A$ in a ferromagnet, which can be a difference of more than two orders of magnitude. Therefore antiferromagnetically ordered spins can change their orientation on a much faster time scale than ferromagnetic ones and this makes antiferromagnets promising materials for the purposes of ultrafast magnetic recording. Indeed, dynamical many-body theory calculations show a possibility of antiferromagnetic dynamics with a time constant of a few femtoseconds only.[9)] Experimentally, the ultrafast dynamics of an antiferromagnet has been an open and intriguing question. The problem however is far from trivial, as there is no straightforward method for the manipulation and detection of spins in antiferromagnetic materials. Therefore, an appropriate mechanism should be found that would deflect the antiferromagnetic moments on a time scale down to femtoseconds, and this change should subsequently be detected on the same time scale. In this article we will describe the details of the technique that allows the detection of the orientation of antiferromagnetically ordered spins with subpicosecond temporal resolution. New approaches for ultrafast laser induced manipulation of antiferromagnetically ordered spins will be presented and a possibility of spin reorientation within 1 ps will be demonstrated. Finally a possible approach for an application of antiferromagnets for the purposes of ultrafast magnetic recording will be suggested.

2. Ultrafast Detection of Antiferromagnetically Ordered Spins

2.1 Linear magnetic birefringence

The most common techniques to study magnetic properties of ferromagnets are based on the measurements of the magneto-optical Faraday or Kerr effects. But since these two phenomena are linear with respect to magnetization, one may expect that they should vanish in antiferromagnetic materials with antiparallel alignment of spins. For the detection of the orientation of spins in antiferromagnets we have chosen linear magnetic birefringence.[13)] This effect is quadratic with

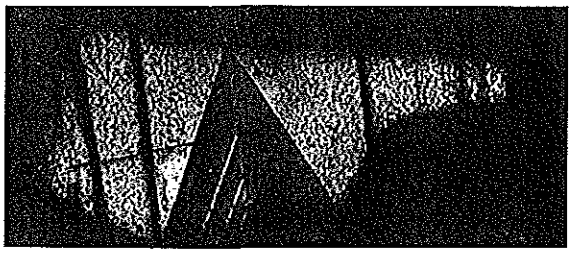

Fig. 1 Antiferromagnetic domains in NiO(111) as they are seen in an ordinary polarization microscope due to the phenomenon of linear magnetic birefringence.[17] Different orientations of antiferromagnetically ordered spins in the domains result in a different birefringence of light in these domains. If one places NiO between two crossed polarizers, the antiferromagnetic domains can be seen as dark (triangle in the centre and vertical stripes) and bright regions.

respect to magnetic moment and thus, in contrast to Faraday and Kerr effects, does not vanish in antiferromagnets.

Linear magnetic birefringence manifests itself in optical anisotropy induced by a magnetic order parameter in general, and the vector of antiferromagnetism, in particular. For example, for light propagating along the z-axis through a birefringent medium, the refractive indices for light polarized along the x-axis and y-axis are different, where Δn_{xy} characterizes the birefringence and is determined by a difference in the diagonal components of the dielectric permittivity tensor ε_{ii}:

$$\Delta n_{xy}=\frac{\varepsilon_{xx}-\varepsilon_{yy}}{2n}$$

If the spins S are ordered an additional contribution to the unperturbed dielectric permittivity tensor appears $\varepsilon_{ii}=\varepsilon_{ii}^{(0)}+\beta_{iikl}S_jS_k$, where $\varepsilon_{ii}^{(0)}$ is unperturbed dielectric permittivity tensor and β_{ijkl} is a polar fourth rank tensor, resulting in an additional change of the birefringence Δn_{xy}. From this phenomenological equation one can see that antiferromagnetically ordered spins can induce an optical axis in a medium and thus make the medium birefringent. The orientation of the induced optical axis is determined by the orientation of the spins and the symmetry of the medium.[14] Thus, the birefringence serves as a direct measure of the orientation of the antiferromagnetically ordered spins S. The sensitivity of the birefringence to the orientation of antiferromagnetically ordered spins has been demonstrated, for instance, in NiO,[5)-17)] where this phenomenon allows one to observe antiferromagnetic domains in a ordinary polarization microscope (see Fig. 1).

2.2 Measurements with subpicosecond temporal resolution

Modern ultrafast (10^{-13} s and faster) laser sources are able to excite a medium at much shorter time scales

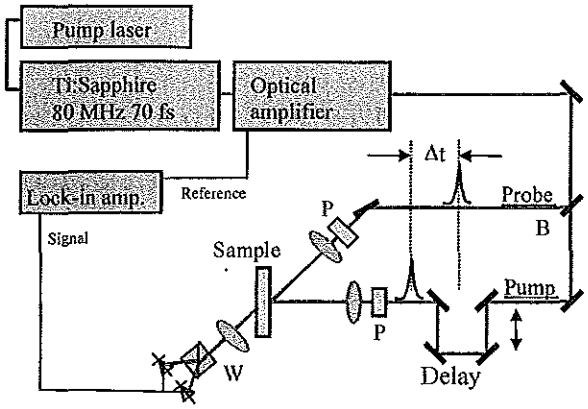

Fig. 2 Scheme of the experimental setup for the study of ultrafast spin dynamics.[18]

than fundamental quantities such as spin precession or spin–lattice relaxation times. Although none of the existing electronic devices can access this time scale, application of such lasers and optical methods allow one to study ultrafast processes in solids, and spin dynamics in particular, with subpicosecond temporal resolution.

For time-resolved measurements we used amplified pulses from a Ti : sapphire laser with repetition rate 1 kHz. Each pulse had an energy of about 2 μJ and a profile close to Gaussian, with a width at half maximum of about 100 fs. The pump and probe beams were linearly polarized with an intensity ratio of about 100. Both beams were focused on the sample to a spot diameter of 50 μm at half maximum for the pump and somewhat smaller for the probe beam. The probe, polarized at 45° with respect to the crystallographic axis in the sample plane, detected the birefringence changes induced by the pump, employing a sensitive two-diode balanced detection scheme. Due to the finite speed of light the position of the spatial delay line determines the time delay between pump and probe pulses at the sample. Varying the time delay between pump and probe pulses one can monitor the magnetic state of a medium with a temporal resolution limited by the pulse width.

3. Ultrafast Laser Manipulation of Antiferromagnetically Ordered Spins

3.1 Photoexcitation of antiferromagnets

Here we describe the process of the excitation of antiferromagnetic materials by subpicosecond laser pulses. For simplicity we consider the case of dielectrics where optical transitions are localized and the photon energy after absorption is transferred to potential energy of electrons and to phonons. For the cases studied in this work the lifetime of the electrons in the excited states does not exceed 100 fs. It means that shortly after photoexcitation the electrons relax back to the ground state and transfer energy to phonons *via*

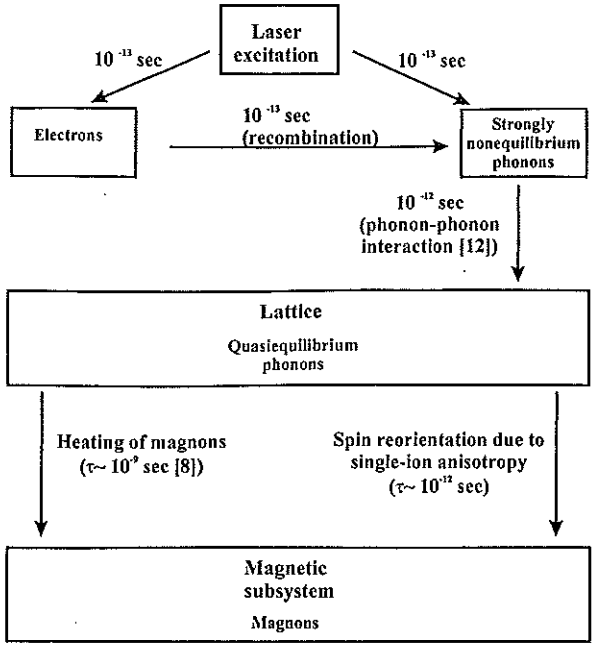

Fig. 3 Scheme that illustrates dynamics of photoexcitation of antiferromagnets

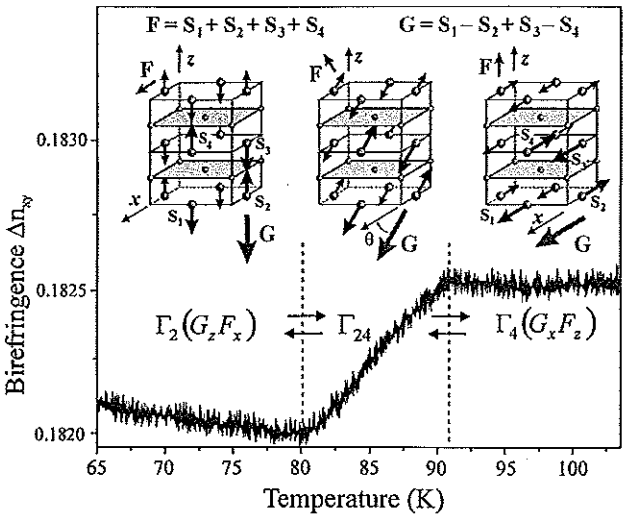

Fig. 4 Magnetic structure and spin-reorientation phase transition in $TmFeO_3$ observed through birefringence measurements.[12]

processes of nonradiative recombination. Thus, the laser pulse within 100 fs creates a number of nonequilibrium phonons. These nonequilibrium phonons will decay via phonon–phonon interaction and the phonons will approach the Bose–Einstein distribution with a temperature higher than before photoexcitation. This decay of nonequilibrium phonons takes at least 1 ps.[19] At this stage the energy delivered by the photoexcitation is accumulated in the lattice, but still not in the magnetic system. However, it has been shown that the speed of heating of magnons due to energy transfer from phonons to magnons is determined by the slow phonon–magnon interaction. For example, in $FeBO_3$ the response of the magnetic system on such photoexcitation takes 2 ns.[8] Therefore, this technique can not serve for purposes of ultrafast magnetic manipulation and other faster methods should be found.

The solution to this problem can be found in the magnetocrystalline anisotropy. Indeed, increase of the lattice temperature can trigger in some antiferromagnets a spin reorientation phase transition. This is equivalent to a rapid change of the magnetic anisotropy that can lead to a reorientation of the spins. The time of spin reorientation in this case is given by the period of homogeneous precession, which can be in antiferromagnets as short as 1 ps.

3.2 Spin-reorientation phase transition in orthoferrites

The rare-earth orthoferrites $RFeO_3$ (where R indicates a rare-earth element) investigated here are known for a strong temperature-dependent anisotropy.[20] These materials crystallize in an orthorombically distorted perovskite structure with four molecular units per unit cell, with a space-group symmetry D_{2h}^{16} (Pbnm). The iron moments order antiferromagnetically, as shown in Fig. 4. There is, in addition, a weak antisymmetric exchange coupling that causes a small canting of the spins on opposite sublattices. The temperature-dependent anisotropy energy has the form: $\Phi(T) = \Phi_0 + K_2(T)\sin^2\theta + K_4\sin^4\theta$, where θ is the angle in the xz-plane between the x-axis and the antiferromagnetic moment G, see Fig. 4, and K_2 and K_4 are the anisotropy constants of second- and fourth-order, respectively. Due to the strong temperature dependence of these anisotropy constants the rare-earth orthoferrites exhibit a continuous spin reorientation over 90 degrees in a specific temperature range. For example, Fe-spins in $TmFeO_3$ align along the z-axis below 80 K and along the x-axis above 90 K. Between 80 K and 90 K the Fe-spins in $TmFeO_3$ exhibit a continuous reorientation. The spin reorientation triggered by the temperature increase in $TmFeO_3$ manifests itself in a changing magnetic linear birefringence as shown in Fig. 4.

3.3 Ultrafast dynamics of spin reorientation

Figure 5 shows the ultrafast dynamics of the birefringence in $TmFeO_3$ for different temperatures. From the figure one can distinguish three distinct processes: 1) the fast exponential changes of the signal in 1 ps after the photoexcitation 2) a slower exponential decay with a characteristic time of about 4 ps, and 3) oscillations of the signal with a frequency of about 80 GHz. According to the scheme in Fig. 3 the first fast exponential recovery of the signal corresponds to decays of the excitation via phonon cascades and the thermalization of the phonon system. In 1 ps after the photoexcitation phonon–phonon interaction sets a new lattice temperature so that the equilibrium anisotropy axis is changed. Under such conditions the antiferromag-

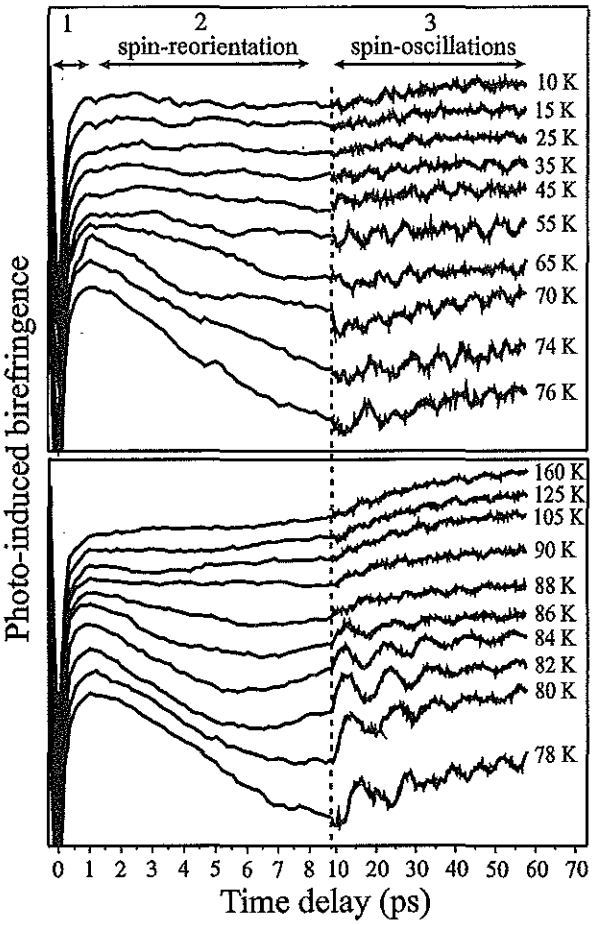

Fig. 5 Dynamics of the laser-induced spin reorientation as it is seen in birefringence measurements. Process 1 corresponds to the period when the new orientation of the easy axis of the magnetic anisotropy is set, process 2 is the reorientation of spins to the new equilibrium, and processes 3 is the oscillations of the spins around the new equilibrium with the frequency of the antiferromagnetic resonance.[12]

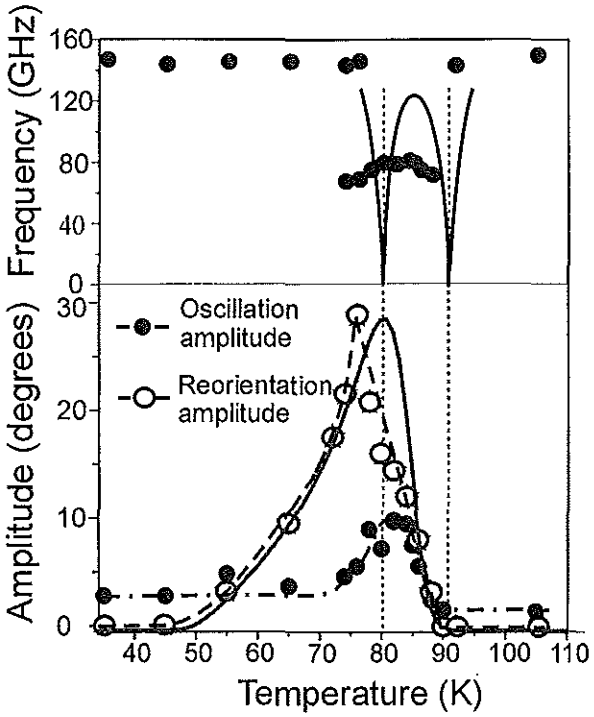

Fig. 6 Temperature dependencies of the amplitudes and frequencies of the observed oscillations, as well as the amplitude of the spin reorientation. Solid black line shows the frequency change at the reorientational transition from earlier measurements.[21] Solid grey line shows the expected response of the sample. The non-zero reorientation amplitude at $T=55$ K corresponds to the instantaneous local laser-induced heating of about 35 K.[12]

netically ordered spins start oscillations around their new equilibrium direction, approaching it due to the damping. This process is marked 2 in the time dependencies of Fig. 5 and has a characteristic time of about 4 ps. This relaxation time corresponds to an antiferromagnetic resonance frequency of 80 GHz. After the initial relaxation, the antiferromagnetic vector oscillates around its new equilibrium (process 3).

The assumption that the exponential decay with characteristic time of 4 ps is related to spin reorientation can be unambiguously proven by the temperature dependence of the amplitude of this exponent. Figure 6 shows that the amplitude grows on approaching the spin reorientation region and drops down in the range between 80 and 90 K. Such thermal behaviour of the amplitude of the detected spin reorientation can be explained by the fact that due to absorption in the sample the temperature profile created by the pump is not uniform. The pump-induced temperature increase as a function of penetration depth x can be expressed as $\Delta T(x)=T_m^* \exp(-kx)$, where k is about 700 1/cm and T_m is about 35 K. The expected response of the sample on such photo-excitation was estimated *via* the integration through the probed volume and shown in Fig. 6 as a function of sample temperature. One can see that calculations and experiment are in very good agreement. Note that such thermal behaviour of the response supposes that somewhere spins get fully reoriented over 90 degrees.

Figure 6 also shows the temperature dependence of the amplitude of the oscillations observed. From the figure one can see that particularly strong oscillations are observable in the range of 80–90 K, *i.e.* in the region of the reorientational transition. This feature clearly indicates that the oscillations should be assigned to spins. Moreover, the frequency of the oscillations is in excellent agreement with the frequency of antiferromagnetic resonance in $TmFeO_3$, that again proves the validity of our interpretation of the experimental results.

The estimated maximum of the reorientation ampli-

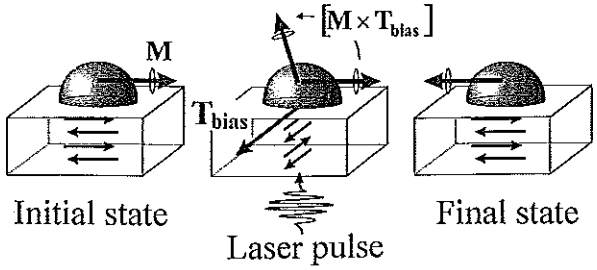

Fig. 7 Ultrafast laser-induced switching of an Ferromagnet/Antiferromagnet bilayer: magnetization of the ferromagnetic layer processes in the exchange field of the antiferromagnet into a final opposite state.[12]

tude of 30 degrees, in fact, is only related to the problem of the instantaneous and homogeneous heating of a bulk sample and can easily be overcome in smaller structures, where such reorientation is of practical importance.

4. Possible Applications of Antiferromagnets in Ultrafast Magnetic Recording

The ultrafast spin reorientation of antiferromagnetically ordered spins has a high potential to be applied for the purposes of ultrafast magnetic recording. For example, in an exchange-coupled Ferromagnet/Antiferromagnet bilayer, the laser-induced reorientation of the antiferromagnetic vector by 90 degrees will trigger, via the exchange coupling, a precession of the ferromagnetic moment into the opposite state. This possible application is illustrated in Fig. 7. The reversal speed of the ferromagnet is determined by a precession of the ferromagnetic spins in the strong interfacial exchange field and does not require any other field. In this structure the spin reorientation time in the ferromagnetic layer is determined not by the intrinsic magnetic anisotropy, but by the interfacial exchange field. The latter, in principle, can be strong and spin reorientation times in the ferromagnetic layer may access the picosecond range. Note that in contrast to hybrid recording, the magnetic anisotropy is not destroyed, but modulated. This results in orders of magnitude faster spin reorientation in ferromagnetic materials. The principle shown in Fig. 7 could be integrated in MRAM when the heating is provided by a current pulse through the ferromagnet. Thus, in addition to increasing the stability of magnetic nanoelements, the antiferromagnetic layer could also play an active role in the ultrafast switching process of ferromagnetic nanostructures.

5. Summary

We have demonstrated that antiferromagnets posses much faster spin dynamics than ferromagnets. For the manipulation of antiferromagnetically ordered spins we have used materials with a strongly temperature dependent magnetic anisotropy, so that an increase of the temperature of the material with a few degrees results in a spin reorientation over 90 degrees. Using ultrafast laser sources one can trigger such spin reorientation within 1 ps, while similar spin reorientations in ferromagnetic materials take hundreds of picoseconds. Therefore antiferromagnets are promising materials for the purposes of ultrafast magnetic recording, and they can play an active role in the ultrafast switching process of ferromagnetic nanostructures. An example of application of antiferromagnetic materials in ultrafast magnetic recording is suggested.

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